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# Intracavity double diode structures with GaInP barrier layers for thermophotonic cooling

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## ABSTRACT

Optical cooling of semiconductors has recently been demonstrated both for optically pumped CdS nanobelts and for electrically injected GaInAsSb LEDs at very low powers. To enable cooling at larger power and to understand and overcome the main obstacles in optical cooling of conventional semiconductor structures, we study thermophotonic (TPX) heat transport in cavity coupled light emitters. Our structures consist of a double heterojunction (DHJ) LED with a GaAs active layer and a corresponding DHJ or a p-n-homojunction photodiode, enclosed within a single semiconductor cavity to eliminate the light extraction challenges. Our presently studied double diode structures (DDS) use GaInP barriers around the GaAs active layer instead of the AlGaAs barriers used in our previous structures. We characterize our updated double diode structures by four point probe IV-measurements and measure how the material modifications affect the recombination parameters and coupling quantum efficiencies in the structures. The coupling quantum efficiency of the new devices with InGaP barrier layers is found to be approximately 10 % larger than for the structures with AlGaAs barriers at the point of maximum efficiency.

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**Keywords:** electroluminescent cooling, quantum efficiency, radiative and non-radiative recombination, III-V semiconductors, double diode structures

## 1. INTRODUCTION

After a few decades with the research spotlight on new and emerging materials such as gallium nitride<sup>1</sup> and antimonides,<sup>2</sup> the mature III-V materials are now gradually recapturing some of their lost research focus. During the past ten years, the fields associated with optical cooling phenomena have witnessed several important advances. In the photoluminescent (PL) cooling (i.e. laser cooling) branch several new records in the cooling of doped glasses have been demonstrated,<sup>3,4</sup> and also the first demonstrations of PL cooling of semiconductor materials have recently taken place.<sup>5</sup> Similarly, in the electroluminescent (EL) cooling branch very low power EL cooling of small bandgap LEDs has been recently reported<sup>6,7</sup> and also the possibilities of thermally enhanced light emission<sup>8,9</sup> have started to attract attention. These advances have already brought us substantially closer to the fundamental limits of high power EL cooling of LEDs as well as high efficiency thermophotonic (TPX) cooling enabling higher coefficient of performance and improved photon transport possibilities. In practice, however, reaching high operation efficiency predicted in theoretical studies<sup>10,11</sup> requires working at higher bias voltages, more efficient light extraction, lower optical losses and higher internal and external quantum efficiencies (IQE and EQE). To reach the high bias operation and high efficiencies, we have recently proposed using a GaAs/AlGaAs based double diode structure (DDS) to overcome the light extraction and efficiency measurement challenges.<sup>12</sup> The proposed design includes an LED and a photodiode embedded in the same optical cavity and

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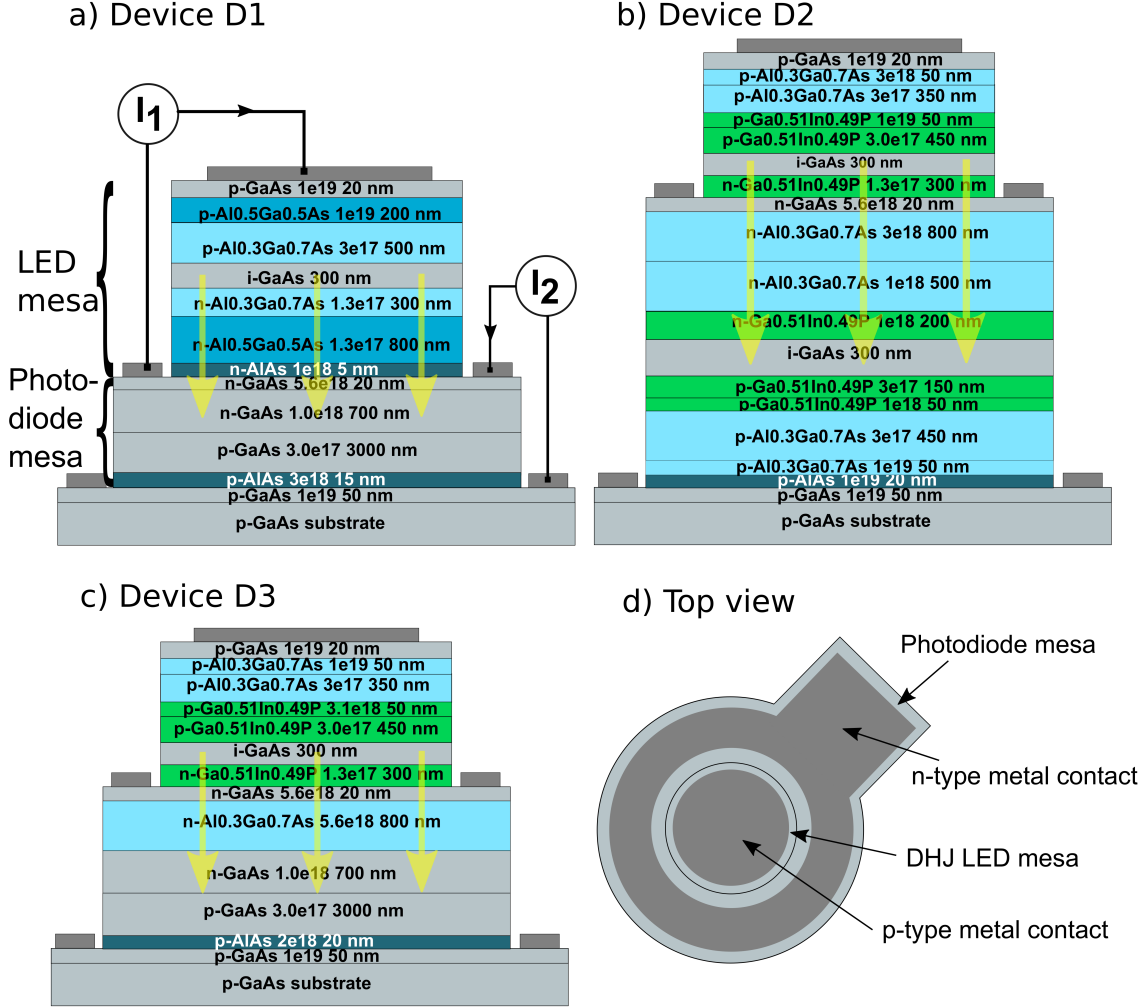


Figure 1. The detailed epitaxial structures and schematics of processed devices a) D1, b) D2, and c) D3. The doping levels are given in  $\text{cm}^{-3}$ . d) A top view of the double mesa structure. The schematic of the IV-IV measurement is also illustrated as an example on the device D1. The coupling quantum efficiency (CQE) of the system is defined as the ratio  $\eta_{\text{CQE}} = I_2/I_1$ .

it aims to provide more insight on the operation of the devices as well as to overcome the light extraction issues by improving the optical interaction between the elements.

Our very first tests of the DDS structures suggested that improving the light emission efficiency to the level needed to demonstrate TPX cooling mainly requires decreasing the losses related with Shockley-Read-Hall and interface/surface recombination, but also highlighted the need to better understand and control other key factors affecting the device performance.<sup>12</sup> Here we report results on our updated DDSs aiming at further improvements in the device efficiency by changes in the selected materials for the epitaxial layers and later on enabling further benefits based on thin film device geometry.

## 2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUES

The devices studied in this work each consist of a double heterojunction (DHJ) LED structures grown on top of homo- or heterojunction photodiodes as illustrated in Fig. 1. The light emitted by the DHJ LED is guided towards the underlying photodiode either directly or after a single reflection from the top contact. Measuring the

current of the photodiode allows a straightforward means to detect the main part of the emitted light without the need to extract it from the high refractive index semiconductor material.

The semiconductor heterostructures used as the base material for the DDS processing are grown by molecular beam epitaxy (MBE) (devices D1 and D2) and metal-organic vapour phase epitaxy (MOVPE) (device D3). The material design is partially based on our previous work with the AlGaAs/GaAs system (Fig. 1a), but in order to reduce the possible interface recombination between GaAs and AlGaAs layers we have introduced GaInP barriers around the GaAs active region in the LED structure (Fig. 1c) as well as in the thin-film variant of the photodiode (Fig. 1b). This modification is expected to reduce the interface recombination rate significantly based on reported interface recombination velocities for GaAs/AlGaAs and GaAs/GaInP interfaces.<sup>13</sup> We have also considered both the doping profile and the material properties at the interfaces to provide an educated guess as a starting point for reducing the potential barriers in actual devices. The structure B is designed to be later processed as a true thin film device where the substrate is removed and replaced by a mirror structure to increase optical confinement and the photodiode absorption efficiency. The epistructure of D3 (Fig. 1c) has GaInP barriers only on the LED side and as a difference to the thin-film device B it has a thick GaAs pn-homojunction photodiode to increase the photodiode efficiency even without substrate removal.

The DDSs were fabricated by several consecutive lithography and wet etching steps first to define the double mesa structure and then to deposit and lift-off n- and p-contacts using standard contact metal combinations. For different material groups and combinations a set of suitable selectively etching acid solutions were used to etch the mesa structure. To estimate the coupling quantum efficiency (CQE) and to gain access to other recombination properties, the main characterization of the devices was done in a IV-IV 4-point probe setup where the IV characteristics of the LED were measured while simultaneously measuring the IV characteristics of the photodiode, as also illustrated in Fig. 1 for D1. We define the CQE of the system as the ratio  $\eta_{\text{CQE}} = I_2/I_1$  where  $I_1(U_1)$  is the current injected through the DHJ LED under bias voltage  $U_1$  and  $I_2(U_2)$  is the photocurrent generated in the lower diode when it is used as a photodetector under short circuit ( $U_2 = 0$ ) or moderate reverse bias conditions ( $U_2 < 0$ ). We also tested the affect of surface passivation treatment with ammonium sulphide by measuring the IV-IV before and after the treatment. In addition to the IV-IV measurements we fabricated modified DDSs where several holes were processed into the top contact metal, so that the light emission and current spreading in the structure could be qualitatively measured by measuring the light emission through the top of the LED.

The four probe IV-IV measurements can also reveal information about the quality and the most important material and device parameters of the DHJ LEDs. The LED recombination processes i.e. the Shockley-Read-Hall (SRH), radiative and Auger recombination create current density components  $J_{\text{SRH}}$ ,  $J_{\text{R}}$  and  $J_{\text{A}}$  that follow the ABC-model<sup>14</sup> which allows to estimate the recombination coefficients  $A$ ,  $B$  and  $C$ , respectively. By using the ABC-model we approximate the  $A$  and  $B$  parameters of our devices according to relations  $A \sim J/[q d n_i \exp(qU/2k_{\text{B}}T)]$  and  $B \sim J/[q d n_i^2 \exp(qU/k_{\text{B}}T)]$ , where  $q$  is the elementary charge,  $d$  is the thickness of the LED active layer,  $n_i$  is the intrinsic carrier concentration,  $U \leq U_1$  is the voltage over the ideal LED,  $k_{\text{B}}$  is the Boltzmann constant and  $T$  is the temperature. We also estimate the SRH recombination coefficient  $A$  by using a relation  $A = J_{\text{SRH}}\sqrt{B}/\sqrt{J_{\text{R}}qd}$  and a literature value for  $B$ . In addition we use the characteristics of the efficiency droop to estimate the internal quantum efficiency (IQE),  $A$ , and  $B$  parameters.<sup>15</sup> All the estimations are described in more detail in our previous work.<sup>12</sup>

### 3. RESULTS AND DISCUSSION

To investigate and characterize the operation of the intracavity systems we analyze the IV-characteristics of the devices in Fig. 1. Figure 2a) shows the current densities of the LED and the photodiode of D3 on a linear scale as functions of the LED bias voltage  $U_1$  and a linear fit to the large current density region where the IV behaviour is dominated by the internal resistance and  $U_2 = 0$ . The corresponding curves for D1 are qualitatively similar and can be found in our previous work.<sup>12</sup> In Fig. 2b) the absolute values of the current densities are shown on a logarithmic scale along with ideal diode curves with ideality factors of 1 and 2 fitted to the purely exponential parts of the IV curves. The figures also show in grey vertical lines the voltage corresponding to the GaAs band gap (1.42 eV) and the voltage corresponding to 80 % of the gap which would allow cooling with  $\eta_{\text{QE}} = 80$  %.

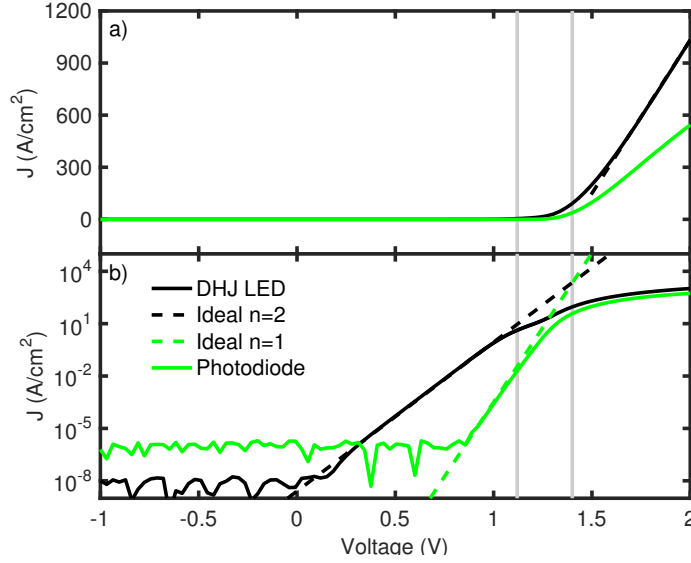


Figure 2. a) The IV curve for a DHJ LED and a photodiode of D3 on a linear scale with a line fitted to the large current density region of the LED, and b) the IV curves of the LED and photodiode as well as ideal diode fits with ideality factors of 1 and 2 on a semilogarithmic scale. The grey vertical lines show the voltages corresponding to  $0.8E_g$  and  $E_g$  energies of GaAs.

Table 1. The recombination parameters for D1, D2 and D3 100  $\mu\text{m}$  mesas estimated with several methods. For the D1 the parameters are obtained for a mesa with an ODR mirror on top.

Parameter	D1	D2	D3
Droop fit method			
$A$ ( $\text{s}^{-1}$ )	$9 \times 10^6$	$2 \times 10^6$	$6 \times 10^6$
$B$ ( $\text{m}^3\text{s}^{-1}$ )	$1 \times 10^{-16}$	$8 \times 10^{-18}$	$2 \times 10^{-17}$
IQE (%)	68	70	81
Approximation method			
$A$ ( $\text{s}^{-1}$ )	$4 \times 10^8$	$4 \times 10^8$	$4 \times 10^8$
$B$ ( $\text{m}^3\text{s}^{-1}$ )	$2 \times 10^{-16}$	$3 \times 10^{-17}$	$2 \times 10^{-16}$
Fitting method			
$A$ ( $\text{s}^{-1}$ )	$7 \times 10^8$	$3 \times 10^8$	$5 \times 10^8$

The fits with ideality factors 1 and 2 in Fig. 2 show that the operation of the DHJ LED is dominated by SRH-like recombination up to voltages of  $\sim 1$  V and current densities of  $\sim 1$  A/cm<sup>2</sup> while the current of the photodiode closely follows the bimolecular form. At voltages exceeding 1 V, however, the IV characteristics of the LED exhibit somewhat unconventional features which was also seen and discussed in our previous work.<sup>12</sup> The IV-IV characteristics are remarkably similar for D1 and D3, however, for D2 the IV curves (not shown) are not entirely consistent with the ideal diode law at this point, presumably due to some issues with the quality of the contacts.

We have estimated the recombination parameters of the materials from the IV-IV measurements using the approximations described in the previous section and considering the condition that  $U \leq U_1$  in the ABC model. These results and the results from droop fit method (using  $C \sim 10^{-30}$  cm<sup>6</sup>s<sup>-1</sup> for GaAs<sup>16</sup>) are shown in Table 1 and are rather similar from device to device, only the droop fit method giving some small differences. The variations between different methods can be explained by their approximate nature, dependence on the Ohmic contact quality and device heating, which are not considered in the estimations and are expected to affect accuracy of the estimates to various degrees. In any case we can conclude that SRH like recombination in the devices is order(s) of magnitude larger than in the best reported devices with SRH limited carrier lifetimes well exceeding 1 $\mu\text{s}$ .<sup>17</sup>

Figure 3 shows the CQE as a function of the DHJ LED current density for D1, D2 and D3 with 100  $\mu\text{m}$

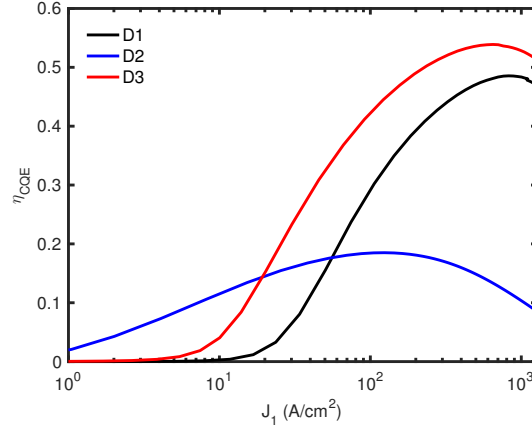


Figure 3. The CQE as a function of the DHJ LED current density for D1, D2 and D3 with 100  $\mu\text{m}$  diameter mesas.

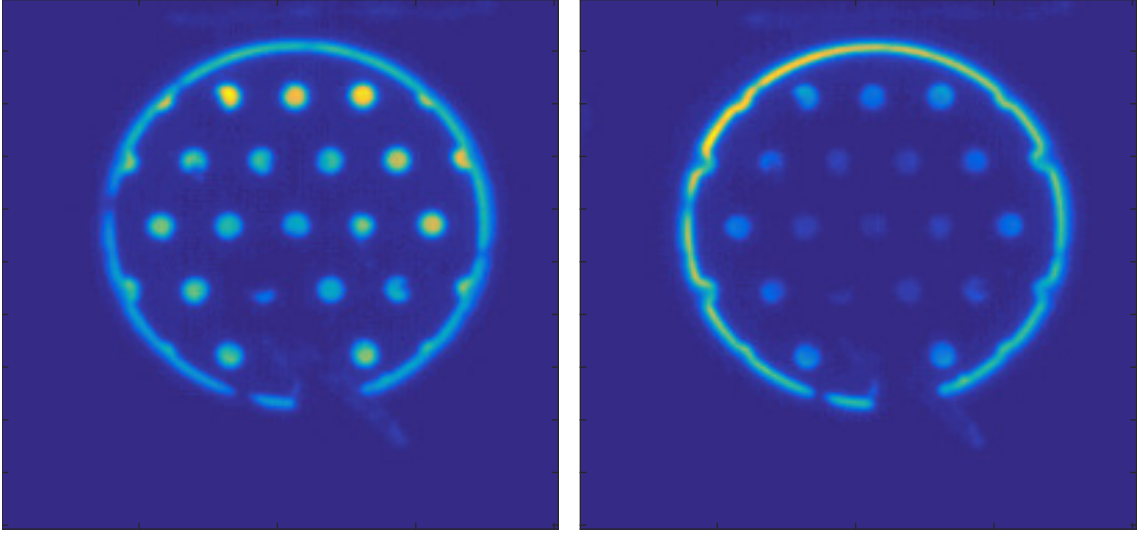


Figure 4. Normalized light emission profile from the device with small openings in the top metal layer at current density 10 mA (left) and 100 mA (right) over a 1 mm diameter mesa. At 10 mA the light emission is relatively uniform over the whole mesa, while for 100 mA the emission is notably stronger at the mesa edges. The contact pad for the n-type layer below the mesa is located on the upper left corner of the image.

diameter mesas. From the data of the D1 we have selected the data for a mesa comparable with D2 and D3, i.e. with no omnidirectional (ODR) mirror on top contact, since this affects the CQE. For D1 and D3 the CQE increases with increasing current up to current densities of  $\sim 300\text{--}800\text{ A/cm}^2$  and then starts to decrease showing a clear tendency of efficiency droop. Device D3 with the GaInP barriers shows the highest CQE, while device D2 has much lower efficiency due to the thinner photodiode active region absorbing a substantially smaller fraction of the light emitted by the LED. In comparison to our earlier DDS structures with AlGaAs barrier layers (D1), for D3 the CQE has increased by  $\sim 10\%$ . At present we cannot fully attribute this only to the structural modifications of the sample, since we would also expect to see a clear change in the  $A$  parameter if this was a direct result of the reduced interface recombination.

Comparing Fig. 3 and 2 reveals an additional challenge for further increases of the CQE. In Fig. 2 D3 starts to shift to series resistance dominated region already at around  $1\text{--}10\text{ A/cm}^2$ , while the efficiency maximum in Fig. 3 is located at  $100\text{--}1000\text{ A/cm}^2$  range. This implies that it will be necessary to reduce the device series resistance, to decrease the peak efficiency current density, or to increase the electron current provided to the LED through the electron-hole generation processes taking place in the solar cell. On a closer inspection, however, each of these objectives are very closely coupled to the objective to further reduce the SRH-like recombination

processes, which is expected to lead to shifting the CQE peak efficiency to smaller carrier densities. This also leads to smaller current density and resistive losses, as well as to increase the efficiency of the charge generation process at the solar cell. Therefore it is expected that it may be possible to solve the current spreading challenge by exactly the same methods that are also needed to increase the overall efficiency of the device.

Finally, to get an idea of the present charge spreading efficiency of the devices, we have imaged the light emission from comparable devices with small openings in the top metal contacts. Fig. 4 shows the normalized luminescence profile through the openings under 10 mA and 100 mA injection current over a large 1 mm diameter mesa. At 10 mA the light emission is relatively uniformly distributed over the mesa while at 100 mA the edges are notably brighter than the openings in the mesa center, highlighting that also surface recombination at the mesa edges may play an important role in determining the device efficiency. In fact, it appears according to our preliminary trials that surface passivation by ammonium sulphide notably decreases the SRH-parameter value estimated using the IV measurements, but in contradiction does not seem to substantially affect the maximum CQE value.

## 4. CONCLUSIONS

To investigate the potential of using intracavity double diode structures for thermophotonic cooling demonstrations, we have fabricated and characterized DDS structures consisting of GaInP barrier layers surrounding the GaAs active layers, alongside with structures with AlGaAs barrier layers. IV-IV four probe measurements were used to estimate and compare the recombination parameters as well as CQE of the devices. The new design with GaInP barriers resulted in approximately 10 % larger CQE compared to devices with AlGaAs barriers, but based on the estimated recombination parameters the origin of the increase does not seem to be directly connected to the lower interface recombination velocities expected in the GaInP devices. In fact, the  $A$  and  $B$  parameters are nearly identical for both barrier types. Despite the clear improvement in the device efficiency, this leaves some of the research questions open, yet providing a significant incentive for future work involving GaInP/GaAs materials.

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## REFERENCES

- [1] Nakamura, S. and Krames, M. R., “History of Gallium Nitride-Based Light-Emitting Diodes for Illumination,” *Proceedings of the IEEE* **101**, 2211–2220 (Oct. 2013).
- [2] Guina, M., Härkönen, A., Korpijärvi, V.-M., Leinonen, T., and Suomalainen, S., “Semiconductor Disk Lasers: Recent Advances in Generation of Yellow-Orange and Mid-IR Radiation,” *Advances in Optical Technologies* **2012**, 1–19 (2012).
- [3] Sheik-Bahae, M. and Epstein, R. I., “Optical refrigeration,” *Nature Photonics* **1**, 693 – 699 (Dec. 2007).
- [4] Epstein, R., Buchwald, M., Edwards, B., Gosnell, T., and Mungan, C., “Observation of laser-induced fluorescent cooling of a solid,” *Nature* **377**(6549), 500–503 (1995).
- [5] Zhang, J., Li, D., Chen, R., and Xiong, Q., “Laser cooling of a semiconductor by 40 Kelvin,” *Nature* **493**, 504–508 (Jan. 2013).
- [6] Santhanam, P., Gray, D. J., and Ram, R. J., “Thermoelectrically pumped light-emitting diodes operating above unity efficiency,” *Phys. Rev. Lett.* **108**, 097403 (Feb 2012).
- [7] Santhanam, P., Huang, D., Ram, R. J., Remennyi, M. A., and Matveev, B. A., “Room temperature thermoelectric pumping in mid-infrared light-emitting diodes,” *Applied Physics Letters* **103**(18), 183513 (2013).
- [8] Xue, J., Zhao, Y., Oh, S.-H., Herrington, W. F., Speck, J. S., DenBaars, S. P., Nakamura, S., and Ram, R. J., “Thermally enhanced blue light-emitting diode,” *Applied Physics Letters* **107**, 121109 (Sept. 2015).



- [9] Oksanen, J. and Tulkki, J., “Thermophotonics: LEDs feed on waste heat,” *Nature Photonics* **9**, 782–784 (Nov. 2015).
- [10] Yen, S.-T. and Lee, K.-C., “Analysis of heterostructures for electroluminescent refrigeration and light emitting without heat generation,” *Journal of Applied Physics* **107**(5), 054513 (2010).
- [11] Yu, S.-Q., Wang, J.-B., Ding, D., Johnson, S., Vasileska, D., and Zhang, Y.-H., “Impact of electronic density of states on electroluminescence refrigeration,” *Solid-State Electronics* **51**(10), 1387 – 1390 (2007). Special Issue: Papers Selected from the {NGC2007} Conference.
- [12] Olsson, A., Tiira, J., Partanen, M., Hakkarainen, T., Koivusalo, E., Tukiainen, A., Guina, M., and Oksanen, J., “Optical Energy Transfer and Loss Mechanisms in Coupled Intracavity Light Emitters,” *IEEE Transactions on Electron Devices* **63**, 3567–3573 (Sept. 2016).
- [13] Levinshtein, M., Rumyantsev, S., and Shur, M., [*Ternary And Quaternary III-V Compounds*], Handbook Series on Semiconductor Parameters, World Scientific Publishing (1999).
- [14] Grundmann, M., [*The Physics of Semiconductors: An Introduction Including Nanophysics and Applications*], Graduate Texts in Physics, Springer International Publishing (2016).
- [15] Kivisaari, P., Riuttanen, L., Oksanen, J., Suihkonen, S., Ali, M., Lipsanen, H., and Tulkki, J., “Electrical measurement of internal quantum efficiency and extraction efficiency of III-N light-emitting diodes,” *Applied Physics Letters* **101**(2), 021113 (2012).
- [16] Strauss, U., Rhle, W. W., and Khler, K., “Auger recombination in intrinsic gaas,” *Applied Physics Letters* **62**(1), 55–57 (1993).
- [17] Bender, D. A., Cederberg, J. G., Wang, C., and Sheik-Bahae, M., “Development of high quantum efficiency GaAs/GaInP double heterostructures for laser cooling,” *Applied Physics Letters* **102**(25), 252102 (4 p.) (2013).